IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

Applicant(s)	:	CHOY and CHANG
U.S. Serial No.	:	09/555,544
For	:	MATERIAL DEPOSITION
Filed	:	August 1, 2000
Examiner	:	John M. Hoffman
Group Art Unit	:	1731

745 Fifth Avenue
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ON AUGUST 27, 2008

REPLY BRIEF

Mail Stop Appeal Brief - Patents
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

This paper is being filed in response to the Examiner's Answer mailed June 27, 2008.

The Examiner's Answer extended the rejection of claims 29-44 under 35 U.S.C. §103(a) as allegedly being unpatentable over Choy et al. (WO 97/21848) in view of Hitachi (JP 56-5337) to claims 52-56 for the first time, and further altered the rejection of claim 51 under 35 U.S.C. §103(a) as allegedly being unpatentable over Hitachi to reference Masahide (JP 64-65040) instead of Japan 62-220376 due to the manner in which the translation is mentioned, as well as the rejection of claim 51 under 35 U.S.C. §103(a) as allegedly being unpatentable over Choy et al. and Blackwell et al. to utilize Masahide (JP 64-65040) instead of Japan 62-220376 due to the manner in which the translation is mentioned. Accordingly, this reply is to function as a Supplemental Appeal Brief, and therefore addresses the revised rejections of claim 51 and the new rejections of 52-56 and all of the rejections addressed in the previous Appeal Brief.

As submitted herewith, the Reply Brief is believed to be in full compliance with 37 C.F.R. §41.37. The Commissioner is hereby authorized to charge any required fee to Deposit Account 50-0230.

INTRODUCTION

This is an Appeal from the December 27, 2005 Final Rejection by the Examiner, finally rejecting claims 29-56 and the Notice of Non-Compliant Appeal Brief dated October 19, 2007. This is also an Appeal from the new ground of rejection of claims 52-56 as identified in the June 27, 2008 Examiner's Answer, as well as the altered grounds of rejection of claim 51, which rejections now utilize Masahide (JP 64-65040) instead of Japan 62-220376.

RELIEF REQUESTED

It is respectfully requested that the rejection of claims 29-56 be reconsidered and withdrawn, and that a Notice of Allowance promptly issue.

REAL PARTY IN INTEREST

The real party in interest is Innovative Materials Processing Technologies Limited, having an address of: 90 Fetter Lane, London, Great Britain, EC4A 1JP.

RELATED APPEALS AND INTERFERENCES

Upon information and belief, the undersigned attorney does not believe that there is any appeal or interference that will directly affect, be directly affected by or have a bearing on the Board's decision in the pending appeal.

STATUS OF THE CLAIMS

Currently pending claims under appeal include claims 29-56. Claims 1-28 have been previously cancelled. Each of claims 29-56 have been rejected as follows:

Claims 45-50 and 52-56 as set forth in Appendix A hereto (Exhibit A), are rejected under 35 U.S.C. §102(b) as being anticipated by Hitachi (JP 56-5337).

Claim 51 as set forth in Appendix A hereto (Exhibit A), is rejected under 35 U.S.C. §103(a) as unpatentable over Hitachi (JP 56-5337) in view of Masahide (JP 64-65040).

Claims 29-50 and 52-56 as set forth in Appendix A hereto (Exhibit A), are rejected under 35 U.S.C. §103(a) as unpatentable over Choy *et al.* (WO 97/21848) in view of Blackwell *et al.* (U.S. Patent 6,312,656).

Claims 29-44 and 52-56 as set forth in Appendix A hereto (Exhbit A), are rejected under 35 U.S.C. §103(a) as unpatentable over Choy *et al.* (WO 97/21848) in view of Hitachi (JP 56-5337).

Claim 51 as set forth in Appendix A hereto (Exhibit A), is rejected under 35 U.S.C. §103(a) as unpatentable over Choy *et al.* (WO 97/21848) in view of Blackwell *et al.* (U.S. Patent 6,312,656) and further in view of Masahide (JP 64-65040).

STATUS OF THE AMENDMENTS

Appellants believe that all the Amendments and papers submitted prior hereto have been entered.

SUMMARY OF THE CLAIMED SUBJECT MATTER

The citations to Figures and Specification locations are provided immediately following elements of independent claims 29, 45 and 51, which are summarized below. However, such citations are provided merely as examples and are not intended to limit the interpretation of the claims or to evidence or create any estoppel. There are three (3) independent claims (claims 29, 45 and 51 on appeal in the instant application, each of which is provided in "non-means" form.

Independent **claim 29** is directed to a method of depositing material on a substrate, comprising the steps of: delivering from a first outlet (60) a stream of droplets of a precursor liquid towards a substrate (50) (see page 1, lines 29-30 of application as filed; Figure 1) applying an electric field between the first outlet (60) and the substrate (50) (see page 1, lines 30-31 of application as filed; Figure 1); and delivering from a second outlet (40) a flow of fuel (see page 2, lines 26-27 of application as filed; Figure 1) about the stream of droplets such as to provide an annular flame combustion region (70) between the first outlet (60) and the substrate (50) (page 3, lines 10-11; Figure 1) through which at least a portion of the stream of droplets passes before reaching the substrate (see page 2, lines 10-11 of the application as filed), whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, to provide the deposited material (see page 2, lines 10-11 of the application as filed). Claims 30-44 depend either directly or indirectly on claim 29 (page 3, lines 9-20 of application as filed).

Independent **claim 45** is directed to an apparatus for depositing material on a substrate as depicted in Figure 1, comprising: a nozzle assembly (10) including a first outlet (60) from which a stream of droplets of a precursor liquid is in use delivered to a substrate (50) (see page 2, lines 5-7 of the application as filed; Figure 1), and a second outlet (40) from which a flow of fuel is in use delivered (see page 2, lines 26-27 of the application as filed; Figure 1) such as to provide an annular flame combustion region (70) (see page 3, lines 9-11 of the application as filed; Figure 1) through which at least a portion of the stream of droplets in use passes before reaching the

substrate (50) (see page 2, lines 10-11 of the application as filed; Figure 1); a precursor supply for supplying a precursor liquid to the nozzle assembly (see page 2, lines 7-8 of the application as filed; Figure 1); an electrical supply (45) for applying an electric field between the first outlet and the substrate (see page 2, lines 8-9 of the application as filed; Figure 1); and a burner for generating the flame of the annular flame combustion region between the first outlet and the substrate (see page 2, lines 9-10 of the application as filed; Figure 1); whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, in the annular flame combustion region to provide the deposited material (see page 2, lines 10-11 of the application as filed). Claims 46-50 and 52-56 depend either directly or indirectly on claim 45 (see page 3, lines 9-20).

Independent **claim 51** is directed to an apparatus for depositing material on a substrate, comprising a nozzle assembly (10) including a first outlet (60) from which a stream of droplets of a precursor liquid is in use delivered to a substrate (50) (see page 1, lines 29-30 of application as filed; Figure 1), and a second outlet (40) from which a flow of fuel (see page 2, lines 26-27 of application as filed; Figure 1) is in use delivered such as to provide an annular flame combustion region (70) (page 3, lines 10-11; Figure 1) through which at least a portion of the stream of droplets in use passes before reaching the substrate (50) (see page 2, lines 10-11 of the application as filed; Figure 1); a mesh (90) (page 3, lines 24-25 of application as filed; Figure 1) disposed between the first outlet (40) and the substrate (50) (see page 3, lines 24-25 of application as filed; Figure 1); a precursor supply for supplying a precursor liquid to the nozzle assembly (10) (see page 2, lines 7-8 of application as filed; Figure 1); an electrical supply (45) for applying an electric field between the first outlet (40) and the substrate (50) (see page 2, lines 8-9 of application as filed; Figure 1); and a burner for generating the flame of the annular flame combustion region (70) between the first outlet (40) and the substrate (50) (see page 2, lines 9-10 of application as filed; Figure 1); whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, in the annular flame combustion region to provide the deposited material, as set forth in claim 51, the third of three independent claims. (see page 3, lines 10-20, 24-25 of application as filed).

GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

Appellants request review of the rejections, specifically:

A. Claims 45-50 and 52-56 as set forth in Appendix A hereto (Exhibit A), are rejected under 35 U.S.C. §102(b) as being anticipated by Hitachi (JP 56-5337).

B. Claim 51 as set forth in Appendix A hereto (Exhibit A), is rejected under 35 U.S.C. §103(a) as unpatentable over Hitachi (JP 56-5337) in view of Japan 62-220376.

C. Claims 29-50 and 52-56 as set forth in Appendix A hereto (Exhibit A), are rejected under 35 U.S.C. §103(a) as unpatentable over Choy *et al.* (WO 97/21848) in view of Blackwell *et al.* (U.S. Patent 6,312,656).

D. Claims 29-44 as set forth in Appendix A hereto (Exhbit A), are rejected under 35 U.S.C. §103(a) as unpatentable over Choy *et al.* (WO 97/21848) in view of Hitachi (JP 56-5337).

E. Claim 51 as set forth in Appendix A hereto (Exhibit A), is rejected under 35 U.S.C. §103(a) as unpatentable over Choy *et al.* (WO 97/21848) in view of Blackwell *et al.* (U.S. Patent 6,312,656) and further in view of Japan 62-220376.

ARGUMENT

THE REJECTION OF ALL CLAIMS UNDER 35 U.S.C. §102 IS OVERCOME

A. Claims 45-50 and 52-56 are rejected under 35 U.S.C. §102(b) as being anticipated by Hitachi (JP 56-5337)

There is no prima facie anticipation between the instant claims and Hitachi.

The instant invention is directed to, *inter alia*, methods of depositing material on a substrate, comprising the steps of delivering from a first outlet a stream of droplets of a precursor liquid towards a substrate; applying an electric field between the first outlet and the substrate; and delivering from a second outlet a flow of fuel about the stream of droplets such as to provide an annular flame combustion region between the first outlet and the substrate through which at least a portion of the stream of droplets passes before reaching the substrate, whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, to provide the deposited material. The present invention is also directed to an apparatus for depositing material on a substrate, comprising: a nozzle assembly including a first outlet from which a stream of droplets of a precursor liquid is in use delivered to a substrate, and a second outlet from which a flow of fuel is in use delivered such as to provide an annular flame combustion region through which at least a portion of the stream of droplets in use passes before

reaching the substrate; a precursor supply for supplying a precursor liquid to the nozzle assembly; an electrical supply for applying an electric field between the first outlet and the substrate; and a burner for generating the flame of the annular flame combustion region between the first outlet and the substrate; whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, in the annular flame combustion region to provide the deposited material.

For a Section 102 rejection to be proper, the single prior art reference must contain <u>all</u> of the elements of the claimed invention, *see Lewmar Marine Inc. v. Barient Inc.,* 3 U.S.P.Q.2d 1766 (Fed. Cir. 1987), and, the single prior art reference must contain an enabling disclosure, *see Chester v. Miller,* 15 U.S.P.Q.2d 1333, 1336 (Fed. Cir. 1990).

It is respectfully submitted that Hitachi does not anticipate the present invention as Hitachi does not teach methods of depositing material on a substrate, comprising the steps of delivering from a first outlet a stream of droplets of a precursor liquid towards a substrate; applying an electric field between the first outlet and the substrate; and delivering from a second outlet a flow of fuel about the stream of droplets such as to provide <u>an annular flame combustion region</u> between the first outlet and the substrate through which at least a portion of the stream of droplets passes before reaching the substrate, whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, to provide the deposited material.

Both the April 13, 2005 Office Action and the December 27, 2005 Office Action state that "Hitachi's apparatus was identical in structure to the claimed apparatus as it included a first central outlet 11', a second outlet 9', a third outlet 7' a precursor supply 1, an electrical supply 21, an annular electrode 23, a positioner 13, and a burner (9', 11' and 7' form a burner face)." It is submitted that this is an incorrect assertion. To the contrary, and as argued in the response filed October 13, 2005 (which argument is set forth here in its entirety), the apparatus of Hitachi cannot result in an annular flame, thereby rendering the present claims patentable over Hitachi. As admitted by the office action, the burner face comprises 9', 11' and 7'. As Applicants previously argued, Hitachi demonstrates a gas entraining a spray of a liquid (1) is ignited and combusted at the mouth of the nozzle (11') through which the gas is delivered. As the nozzle (11') is an open, tubular section, the resulting flame manifestly cannot be annular as now required by the claimed invention. Furthermore, both the April 13, 2005 Office Action and the December 27, 2005 Office Action's characterization of the burner face supports Applicant's

contention that the resulting flame is not annular as a burner face necessarily implies a flame across the entire face - in this instance, across 9’, 11’, and 7’ - such that the flame cannot be annular.

In response to the argument set forth by the Applicant in the October 13, 2005 Response, the Examiner stated that Applicant's arguments based on drawings are not well taken. Applicants submit that the burden was on the Examiner to point to some part of Hitachi which concretely described an annular flame, which the Examiner failed to do.

The Examiner has repeatedly indicated that the cited reference “intends” the use of an annular flame, although no such specific teaching is present in the reference. Appellant reiterates that what the reference may or may not “intend” is different from that which is “taught” by a reference. For a §102 rejection to be proper, the reference must **teach** each and every element of the claims. Suggested “intentions” of a reference, as determined solely by the Examiner, are not sufficient.

Furthermore, the Examiner’s argument presupposes that one flame is the same as any other flame; according to the Examiner, any “flame” necessarily anticipates or renders obvious an “annular” flame. Appellants disagree. Enclosed herewith as Exhibit B1 is Charojrochkul, et al.’s “Flame assisted vapour deposition of cathode for solid oxide fuel cells. 1. Microstructure control from processing parameters.” (Journal of the European Ceramic Society, 2004, 24:2515-2526). Choarojrochkul et al. includes Kwang-Leong Choy, the present inventor, as one of the authors. At page 2517, second paragraph, it is noted that “Three different flame configurations were produced by changing the deposition parameters….The shape of the combustion zone was later found to have a **marked effect** on the microstructure of the deposited film.” (Emphasis added).

That is, post-filing, peer-reviewed references have demonstrated that significant differences in the deposited material occurs when the flame configurations are altered. The shape of the flame, i.e., an annular flame vs. a continuous flame, is clearly applicable to the flame configuration, such that an alteration between the flame shapes would significantly alter the material deposition. One of skill in the art would certainly be aware of the relationship between flame configurations (including shape) and material deposition properties, and therefore would not attempt to alter the exact flame configuration of a reference, including the flame

shape. Thus, one of skill in the art would not read a reference that **does not specifically describe** an annular flame as teaching or referring to an annular flame.

As the pending claims require the presence of an annular flame and the Examiner has failed to specifically indicate where Hitachi teaches an annular flame, the applicant maintains that Hitachi does not disclose the generation of an annular flame combustion region as alleged by the Examiner.

THE REJECTION OF ALL CLAIMS UNDER 35 U.S.C. §103 IS OVERCOME

The instant invention is directed to, *inter alia*, methods of depositing material on a substrate, comprising the steps of delivering from a first outlet a stream of droplets of a precursor liquid towards a substrate; applying an electric field between the first outlet and the substrate; and delivering from a second outlet a flow of fuel about the stream of droplets such as to provide an annular flame combustion region between the first outlet and the substrate through which at least a portion of the stream of droplets passes before reaching the substrate, whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, to provide the deposited material. The present invention is also directed to apparatus for depositing material on a substrate, comprising: a nozzle assembly including a first outlet from which a stream of droplets of a precursor liquid is in use delivered to a substrate, and a second outlet from which a flow of fuel is in use delivered such as to provide an annular flame combustion region through which at least a portion of the stream of droplets in use passes before reaching the substrate; a precursor supply for supplying a precursor liquid to the nozzle assembly; an electrical supply for applying an electric field between the first outlet and the substrate; and a burner for generating the flame of the annular flame combustion region between the first outlet and the substrate; whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, in the annular flame combustion region to provide the deposited material.

In order to ground an obviousness rejection, there must be some teaching which would have provided the necessary incentive or motivation for modifying the reference's teaching. *In re Laskowski*, 12 U.S.P.Q. 2d 1397, 1399 (Fed. Cir. 1989); *In re Obukowitz*, 27 U.S.P.Q. 2d 1063 (B.P.A.I. 1993). Further, "obvious to try" is not the standard under 35 U.S.C. §103. *In re*

Fine, 5 U.S.P.Q. 2d 1596, 1599 (Fed. Cir. 1988). And as stated by the Court in *In re Fritch*, 23 U.S.P.Q. 2d 1780, 1783-1784 (Fed. Cir. 1992): "The mere fact that the prior art may be modified in the manner suggested by the Examiner does not make the modification obvious unless the prior art suggests the desirability of the modification." Also, the Examiner is respectfully reminded that for the Section 103 rejection to be proper, both the suggestion of the claimed invention and the expectation of success must be founded in the prior art, and not Applicants' disclosure. *In re Dow*, 5 U.S.P.Q.2d 1529, 1531 (Fed. Cir. 1988).

B. Claim 51 is rejected under 35 U.S.C. §103(a) as unpatentable over Hitachi (JP 56-5337) in view of Masahide

There is no prima facie obviousness between the instant claim and Hitachi in combination with Masahide.

It is respectfully submitted that Hitachi in combination with Masahide does not render obvious the apparatus of the present invention, having a nozzle assembly including a first outlet from which a stream of droplets of a precursor liquid is in use delivered to a substrate, and a second outlet from which a flow of fuel is in use delivered such as to provide an annular flame combustion region through which at least a portion of the stream of droplets in use passes before reaching the substrate; a mesh disposed between the first outlet and the substrate; a precursor supply for supplying a precursor liquid to the nozzle assembly; an electrical supply for applying an electric field between the first outlet and the substrate; and a burner for generating the flame of the annular flame combustion region between the first outlet and the substrate; whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, in the annular flame combustion region to provide the deposited material.

Both the April 13, 2005 Office Action and the December 27, 2005 Office Action alleged that claim 51 was obvious over Hitachi in view of Masahide as Hitachi taught an apparatus having a first central outlet 11', a second outlet 9', a third outlet 7' a precursor supply 1, an electrical supply 21, an annular electrode 23, a positioner 13, and a burner (9', 11' and 7' form a burner face) and that Japan 62-220376 taught the presence of a wire-mesh electrode. Applicants disagree with this assertion.

To the contrary, and as argued in the response filed October 13, 2005 (which arguments are set forth herein), the apparatus of Hitachi cannot result in an annular flame, thereby rendering the present claims patentable over Hitachi. As admitted by the office action, the burner face

comprises 9', 11' and 7'. As Applicants previously argued, Hitachi demonstrates a gas entraining a spray of a liquid (1) is ignited and combusted at the mouth of the nozzle (11') through which the gas is delivered. As the nozzle (11') is an open, tubular section, the resulting flame manifestly cannot be annular as now required by the claimed invention. Furthermore, both the April 13, 2005 Office Action and the December 27, 2005 Office Action's characterization of the burner face supports Applicant's contention that the resulting flame is not annular as a burner face necessarily implies a flame across the entire face - in this instance, across 9', 11', and 7' - such that the flame cannot be annular.

Again, the Examiner has repeatedly indicated that the cited reference "intends" the use of an annular flame, although no such specific teaching is present in the reference. Appellant reiterates that what the reference may or may not "intend" is different from that which is "taught" by a reference. For a §103 rejection to be proper, the reference must teach or suggest every element of the claims. Suggested "intentions" of a reference, as determined solely by the Examiner and without actual support in the reference, are not sufficient.

Furthermore, the Examiner's argument presupposes that one flame is the same as any other flame; according to the Examiner, any "flame" necessarily anticipates or renders obvious an "annular" flame. Appellants disagree. As described previously, Exhibit B1, Charojrochkul, et al.'s states at page 2517, second paragraph, that "Three different flame configurations were produced by changing the deposition parameters....The shape of the combustion zone was later found to have a **marked effect** on the microstructure of the deposited film." (Emphasis added).

That is, post-filing, peer-reviewed references have demonstrated that significant differences in the deposited material occurs when the flame configurations are altered. The shape of the flame, i.e., an annular flame vs. a continuous flame, is clearly applicable to the flame configuration, such that an alteration between the flame shapes would significantly alter the material deposition. One of skill in the art would certainly be aware of the relationship between flame configurations (including shape) and material deposition properties, and therefore would not attempt to alter the exact flame configuration of a reference, including the flame shape. Thus, one of skill in the art would not read a reference that **does not specifically describe** an annular flame as teaching or suggesting an annular flame.

As argued in the October 13, 2005 Response (which arguments are set forth in their entirety herein), the mesh electrode of Masahide serves to create a corona discharge between two

electrodes. This is in contrast to the present invention, which already contains an electric field which is generated from high voltage source 45, and exists between the nozzle assembly 10 and substrate 50. Rather than acting as the source of the electric field in the present invention, and indeed, the mesh of the present invention is not related to the presence of the electric field in any sense, the mesh instead serves to assist in removing soot from the flame. The Examiner indicates that it would have been obvious to use a wire-mesh electrode like that of Masahide with the arrangement of Hitachi "because a mesh would have been equivalent to the electrode of Hitachi." This assertion has no basis, however, as there is no motivation for one of skill in the art to add a mesh electrode to an apparatus which already contains means for generating an electric field.

In summary, it is submitted that a person skilled in the art would not have contemplated modifying the apparatus or method of Hitachi in accordance with the teaching of Masahide to use a wire mesh electrode, and furthermore such a combination fails to teach an annular flame, and, notwithstanding any such modification of Hitachi, the claimed invention is clearly distinguished over the disclosures of Hitachi and Masahide when taken in combination.

C. Claims 29-50 and 52-56 are rejected under 35 U.S.C. §103(a) as unpatentable over Choy *et al.* <u>(WO 97/21848) in view of Blackwell *et al.* (U.S. Patent 6,312,656)</u>

There is no prima facie obviousness between the instant claims and Choy et al. in combination with Blackwell et al.

It is respectfully submitted that the combination of Choy *et al.* and Blackwell *et al.* do not render the present invention obvious, i.e. Choy *et al.* and Blackwell *et al.* do not combine to provide methods of depositing material on a substrate, comprising the steps of delivering from a first outlet a stream of droplets of a precursor liquid towards a substrate; applying an electric field between the first outlet and the substrate; and delivering from a second outlet a flow of fuel about the stream of droplets such as to provide <u>an annular flame combustion region</u> between the first outlet and the substrate through which at least a portion of the stream of droplets passes before reaching the substrate, whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, to provide the deposited material. Nor does such a combination provide an apparatus for depositing material on a substrate, comprising: a nozzle assembly including a first outlet from which a stream of droplets of a precursor liquid is in use delivered to a substrate, and a second outlet from which a flow of fuel is in use delivered

such as to provide an annular flame combustion region through which at least a portion of the stream of droplets in use passes before reaching the substrate; a precursor supply for supplying a precursor liquid to the nozzle assembly; an electrical supply for applying an electric field between the first outlet and the substrate; and a burner for generating the flame of the annular flame combustion region between the first outlet and the substrate; whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, in the annular flame combustion region to provide the deposited material. Specifically, the rejection relies on Blackwell *et al.* as disclosing the generation of an annular flame combustion region. Applicants respectfully assert that this is not a correct reading of Blackwell *et al.*

Each of the March 5, 2002, November 12, 2002, June 9, 2003, June 11, 2004, April 13, 2005 and December 27, 2005 Office Actions have stated that Choy *et al.* does not show, "generating a flame from a burner coaxial with the droplet outlet," and that Blackwell *et al.* shows, "combustion processes and apparatus for atomized liquid reactants wherein atomization can be done electrostatically." December 27, 2005 Office Action at 5. Additionally, the Office Actions continued that while Choy *et al.* "does not show the claimed sequence of annular gas jets," Blackwell *et al.* shows the "use of inert shield gas between liquid precursor droplets and an outer, annular fuel gas jet." December 27, 2005 Office Action at 5.

The Examiner was apparently alleging that fuel is delivered through an annular channel, namely, the outermost channel (47), and, as such, the flame combustion region (23) must necessarily be annular. It is submitted that this allegation has no basis.

While the Office Actions were correct in stating that Blackwell *et al.* shows combustion processes, it was respectfully submitted that these combustion processes do not include the annular fuel gas jet alleged in the Office Actions.

Rather, Blackwell *et al.* demonstrates the use of a single or multi-jet fuel source which gives rise to a single, continuous flame area. The Examiner was respectfully invited to review figures 2-4 of Blackwell *et al.*, especially figure 4, which demonstrates that the burner (40) includes a number of concentric channels, but that the concentric channels provide a single, continuous flame area, <u>not</u> an annular flame combustion region as stated in the claims of the present invention.

In response to Appellant's comments, the November 12, 2002 Office Action alleged that the disclosure of Figure 4 incorrectly represents the flame combustion region (23) and that the

shape of the flame in Figure 4 could not be relied upon. Specifically, the Office Action alleged that fuel is delivered through an annular channel, namely, the outermost channel (47), and, as such, the flame combustion region (23) must necessarily be annular. It is respectfully submitted that this allegation has no basis.

As Appellants stated in the May 12, 2003 Response (which was accompanied by a Request for Continued Examination, the arguments of which are provided herein) Appellants acknowledge that Blackwell *et al*. discloses the delivery of fuel through an annular outlet, this being the outlet defined by the outermost channel (47), but the outermost channel (47) is a frusto-conical channel and not a cylindrical channel. Blackwell *et al* makes no disclosure of the channels (43-47) being cylindrical channels as alleged by the Office Action, but rather that the channels (43-47) are concentric (column 9, lines 6 and 7). Concentricity merely defines that the channels (43-47) have a common center. The outermost channel (47), in being a frusto-conical channel, manifestly cannot provide for the generation of an annular flame combustion region as alleged in the Office Action.

In this regard, it is submitted that the teaching of Figure 4 is quite clear, in disclosing the generation of a single, continuous flame combustion region (23) from a frusto-conical channel (47). There is nothing in the teaching of Blackwell *et al.* to suggest that Figure 4 incorrectly represents the flame combustion region (23). On the contrary, the inwardly-directed frusto-conical channel (47) as embodied can only provide a single, continuous flame combustion region (23), and, as such, the representation of Figure 4 is submitted to be accurate. Indeed, given that the purpose of the flame combustion region (23) is to provide a conversion site for converting the precursor material into soot particles at the burner face (53) (column 9, lines 13 to 16), it is submitted that the provision of other than a single, continuous flame combustion region (23) would not achieve this purpose. The Examiner is manifestly impermissibly performing a hindsight analysis of the prior art in attempting selectively to disregard the teaching of Figure 4 of Blackwell *et al.*

In support of this allegation, the Office Action is relying on the disclosure at column 9, lines 17 to 20, which discloses that "An inert gas, … is delivered through channel 44 to inhibit reaction of liquid feedstock and soot build-up on burner face 53." The Office Action considers this disclosure to support his allegation that the flame combustion region (23) is away from the burner face (53), and apparently in an annular region defined by the outermost channel (47).

It is, however, submitted that this teaching referenced in the Office Action in fact contrarily demonstrates that the flame combustion region (23) extends across the burner face (53), particularly the central region thereof which includes the atomizer (41) from which feedstock is delivered. The purpose of delivering an inert gas through an inner channel, namely, inner channel (44), is expressly recited as being to "inhibit reaction of the liquid feedstock and soot build-up on burner face (53)." It is submitted that such inhibition at the burner face (53) is required for the very reason that the flame combustion region (23) extends thereover, as, otherwise, inhibition would be unnecessary, and, as such, and contrary to the Office Action's allegation, this disclosure is not to the development of an annular flame combustion region (23).

The Examiner is further alleging that the applicant has failed to identify any teaching in Blackwell *et al.*, other than the representation of the flame combustion region (23) in Figure 4, that the flame combustion region (23) is a single, continuous region.

Notwithstanding that the identification of any further teaching should not be required, given the clear disclosure of Figure 4, the applicant has already identified further clear teaching in Blackwell *et al.* to the flame combustion region (23) being a single, continuous region.

As set out hereinabove, the outermost channel (47) is an inwardly-directed frusto-conical channel, and such an inwardly-directed frusto-conical channel (47) as embodied can only provide a single, continuous flame combustion region (23). The generation of a single, continuous flame combustion region (23) results from the outermost channel (47) being a frusto-conical channel.

Indeed, Blackwell *et al.* further discloses (column 8, line 67 to column 9, line 3) that the atomizer (41) "injects very finely atomized liquid reactant particles **into** flame 23." For such injection to occur into the flame combustion region (23), the flame combustion region (23) has to extend over the atomizer (41), and, as such, the flame combustion region (23) cannot be annular as alleged by the Office Action.

Furthermore, Blackwell *et al.* discloses (column 9, lines 13 to 17) that "The area proximate to the burner face 53 and flame 23 thus acts as a conversion site for converting liquid projections 42 into soot reactant particles." As the liquid projections (42) are created at the outlet of the atomizer (41), and the flame combustion region (23) is required to act as the conversion site for converting the liquid projections (42) into soot reactant particles, and the conversion site is required to be proximate the burner face (53), the flame combustion region

(23) manifestly has to extend over the atomizer (41), and, as such, cannot be annular as alleged in the Office Action. If the flame combustion region (23) were annular as alleged in the Office Action, the stated conversion of the liquid projections (42) would manifestly not occur proximate the burner face (53).

The June 9, 2003 Office Action maintained the previous rejection, and again reiterated the opinion that Blackwell *et al*. teaches an annular flame region.

Appellants presented arguments in the December 9, 2003 Response (which arguments are provided herein) again stating that Blackwell et al. does not describe an annular flame combustion region, as discussed herein above.

Again, Applicants acknowledge that Blackwell *et al.* describes the delivery of fuel through an annular <u>outlet</u>, this being the outlet defined by the outermost channel (47), but the outermost channel (47) is a frusto-conical channel and not a cylindrical channel. Blackwell *et al.* makes no disclosure of the channels (43-47) being cylindrical channels as alleged by the Examiner, but rather the channels (43-47) are concentric (column 9, lines 6 and 7). Concentricity merely defines that the channels (43-47) have a common center, and is not necessarily indicative of an annular flame combusion region. Furthermore, and perhaps most importantly, the outermost channel (47), in being a frusto-conical channel, manifestly cannot provide for the generation of an annular flame combustion region as alleged by the Examiner.

Furthermore, the Examiner again alleged that the disclosure of Figure 4 of Blackwell *et al.* incorrectly represents the flame combustion region (23), and cannot be relied upon. Appellants respectfully disagree, and maintain that Figure 4 is not ambiguous; the flame depicted in Figure 4 is clearly drawn as a continuous flame.

In this regard, it is submitted that the teaching of Figure 4 is quite clear, in disclosing the generation of a single, continuous flame combustion region (23) from a frusto-conical channel (47). There is nothing in the teaching of Blackwell *et al.* to suggest that Figure 4 incorrectly represents the flame combustion region (23). On the contrary, the inwardly-directed frusto-conical channel (47) as embodied can only provide a single, continuous flame combustion region (23), and, as such, the representation of Figure 4 is submitted to be accurate. Indeed, given that the purpose of the flame combustion region (23) is to provide a conversion site for converting the precursor material into soot particles at the burner face (53) (column 9, lines 13 to 16), it is submitted that the provision of other than a single, continuous flame combustion region (23)

would not achieve this purpose. The Examiner is manifestly impermissibly performing a hindsight analysis of the prior art in attempting selectively to disregard the teaching of Figure 4 of Blackwell *et al.*

Again, disregarding the arguments previously filed by Appellants, the Examiner further alleged that no teaching in Blackwell *et al.*, other than the representation of the flame combustion region (23) in Figure 4, had been pointed to by Appellants in demonstrating that the flame combustion region (23) of Blackwell *et al.* is a single, continuous region.

Notwithstanding that the identification of any further teaching should not be required, given the clear disclosure of Figure 4, the applicant has already identified further clear teaching in Blackwell *et al* to the flame combustion region (23) being a single, continuous region.

Firstly, as set out hereinabove, the outermost channel (47) is an inwardly-directed frusto-conical channel, and such an inwardly-directed frusto-conical channel (47) as embodied can only provide a single, continuous flame combustion region (23). The generation of a single, continuous flame combustion region (23) results from the outermost channel (47) being a frusto-conical channel.

Secondly, Blackwell *et al.* further discloses (column 8, line 67 to column 9, line 3) that the atomizer (41) "injects very finely atomized liquid reactant particles **into** flame 23" (emphasis added). For such injection to occur into the flame combustion region (23), the flame combustion region (23) has manifestly to extend over the atomizer (41), and, as such, cannot be annular as alleged by the Examiner.

Thirdly, Blackwell *et al.* discloses (column 9, lines 13 to 17) that "The area proximate to the burner face 53 and flame 23 thus acts as a conversion site for converting liquid projections 42 into soot reactant particles.". As the liquid projections (42) are created at the outlet of the atomizer (41), and the flame combustion region (23) is required to act as the conversion site for converting the liquid projections (42) into soot reactant particles, and the conversion site is required to be proximate the burner face (53), the flame combustion region (23) manifestly has to extend over the atomizer (41), and, as such, cannot be annular as alleged by the Examiner. If the flame combustion region (23) were annular as alleged by the Examiner, the stated conversion of the liquid projections (42) would manifestly not occur proximate to the burner face (53).

Fourthly, the disclosure at column 9, lines 17 to 20, which discloses that "An inert gas, … is delivered through channel 44 to inhibit reaction of the liquid feedstock and soot build-up on

burner face 53", demonstrates that the flame combustion region (23) extends across the burner face (53), particularly the central region thereof which includes the atomizer (41) from which feedstock is delivered. The purpose of delivering an inert gas through an inner channel, namely, inner channel (44), is expressly recited as being to "inhibit reaction of the liquid feedstock and soot build-up on burner face 53." It is submitted that such inhibition at the burner face (53) is required for the very reason that the flame combustion region (23) extends thereover, as, otherwise, inhibition would be unnecessary.

Furthermore, the Examiner still alleges that a person skilled in the art would have been motivated to modify the apparatus and method of Choy *et al.* in accordance with the teaching of Blackwell *et al.* to utilize a combustion flame as the heating means for converting the precursor material.

Similar allegations have been maintained by the Examiner in the June 11, 2004 Office Action and the April 13, 2005 Office Action (which referred to the comments in the June 11, 2004 Office Action) wherein the Examiner alleged that the teaching of an annular flame was inherent with Blackwell *et al.* Applicants have maintained that such a teaching is not provided by Blackwell *et al.*, inherently or otherwise, as the Blackwell *et al.* itself, and specifically Figure 4 has demonstrated otherwise.

Appellants maintain that a person skilled in the art would manifestly not have been so motivated to read Blackwell *et al.* as inherently teaching an annular flame, and at no time has the applicant acquiesced to the Examiner's allegation in this regard.

Choy *et al.* is directed to an apparatus and method which requires an increasing temperature gradient between the outlet (5) and the substrate (14), as disclosed in the summary of the invention at page 1, lines 22 to 30, particularly lines 29 and 30. It is important to recognize that Choy *et al.* does not merely require that an increased temperature be maintained between the outlet (5) and the substrate (14), but rather that an increasing temperature gradient be provided, and more specifically that the increasing temperature gradient be such that the precursor material undergoes de-composition and/or chemical reaction on or in very close proximity to the surface of the substrate (14), as summarized at page 8, lines 22 to 31. The provision of such a heating regime represents the main thrust of the teaching of Choy *et al.*, and, indeed, is disclosed at, for example, page 7, lines 22 and 23 as being the principle of the deposition technique of Choy *et al.*

Given that the teaching of Choy *et al.* is to a deposition technique which essentially requires such a heating regime, it is submitted that a person skilled in the art would have had no possible motivation to contemplate modifying the apparatus or method of Choy *et al.* in a manner which would have been contrary to the very teaching thereof, that is, to provide a heating regime which did not provide an increasing temperature gradient from the outlet (5) to the substrate (14), but contrarily provided for conversion of the precursor material at the outlet (5) and away from the substrate (14).

This notwithstanding, the applicant maintains that Blackwell *et al.* does not disclose the generation of an annular flame combustion region as alleged by the Examiner.

As described above, the Examiner has repeatedly indicated that Blackwell teaches the use of an annular flame, although no such specific teaching is present in the reference, and indeed the reference teaches away from an annular flame as described above. Appellant reiterates that what the reference may or may not "intend" is different from that which is "taught" by a reference. For a §103 rejection to be proper, the reference must teach or suggest every element of the claims. Suggested "intentions" of a reference, as determined solely by the Examiner and without actual support in the reference, are not sufficient.

Furthermore, the Examiner's argument presupposes that one flame is the same as any other flame; according to the Examiner, any "flame" necessarily anticipates or renders obvious an "annular" flame. Appellants disagree. As described previously, Exhibit B1, Charojrochkul, et al.'s states at page 2517, second paragraph, that "Three different flame configurations were produced by changing the deposition parameters….The shape of the combustion zone was later found to have a **marked effect** on the microstructure of the deposited film." (Emphasis added).

That is, post-filing, peer-reviewed references have demonstrated that significant differences in the deposited material occur when the flame configurations are altered. The shape of the flame, i.e., an annular flame vs. a continuous flame, is clearly applicable to the flame configuration, such that an alteration between the flame shapes would significantly alter the material deposition. One of skill in the art would certainly be aware of the relationship between flame configurations (including shape) and material deposition properties, and therefore would not attempt to alter the exact flame configuration of a reference, including the flame shape. Thus, one of skill in the art would not read a reference that **does not specifically describe** an annular flame as teaching or suggesting an annular flame.

In summary, it is submitted that a person skilled in the art would not have contemplated modifying the apparatus or method of Choy *et al.* in accordance with the teaching of Blackwell *et al.* to utilize a combustion flame, and, notwithstanding any such modification of Choy *et al.*, the claimed invention is clearly distinguished over the disclosures of Choy *et al.* and Blackwell *et al.* when taken in combination.

D. Claims 29-44 and 52-56 are rejected under 35 U.S.C. §103(a) as unpatentable over Choy *et al.* (WO 97/21848) in view of Hitachi (JP 56-5337)

There is no prima facie obviousness between the instant claims and Choy et al. in combination with Hitachi.

It is respectfully submitted that Choy *et al*. in combination with Hitachi does not render obvious the present invention of methods of depositing material on a substrate, comprising the steps of delivering from a first outlet a stream of droplets of a precursor liquid towards a substrate; applying an electric field between the first outlet and the substrate; and delivering from a second outlet a flow of fuel about the stream of droplets such as to provide <u>an annular flame combustion region</u> between the first outlet and the substrate through which at least a portion of the stream of droplets passes before reaching the substrate, whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, to provide the deposited material.

The April 13, 2005 and December 27, 2005 Office Actions indicated that "Choy *et a*. did not teach an annular flame" and that "As clearly indicated in Figure 1 of Hitachi, the flame '16 is annular". Applicants agree that Choy *et al*. does not teach an annular flame, however, Applicants disagree with the assertion that Hitachi teaches an annular flame, and have never waived from this position.

Both the April 13, 2005 Office Action and the December 27, 2005 Office Action state that “Hitachi’s apparatus was identical in structure to the claimed apparatus as it included a first central outlet 11’, a second outlet 9’, a third outlet 7’ a precursor supply 1, an electrical supply 21, an annular electrode 23, a positioner 13, and a burner (9’, 11’ and 7’ form a burner face).” It is submitted that this is an incorrect assertion. To the contrary, and as argued in the response filed October 13, 2005 (which arguments are provided herien), the apparatus of Hitachi cannot result in an annular flame, thereby rendering the present claims patentable over Hitachi.

As admitted by the office action, the burner face comprises 9', 11' and 7'. As Applicants previously argued, Hitachi demonstrates a gas entraining a spray of a liquid (1) is ignited and combusted at the mouth of the nozzle (11') through which the gas is delivered. As the nozzle (11') is an open, tubular section, the resulting flame manifestly cannot be annular as now required by the claimed invention. Furthermore, both the April 13, 2005 Office Action and the December 27, 2005 Office Action's characterization of the burner face supports Applicant's contention that the resulting flame is not annular as a burner face necessarily implies a flame across the entire face - in this instance, across 9', 11', and 7' - such that the flame cannot be annular.

Again, the Examiner has repeatedly indicated that the cited reference "implies" the use of an annular flame, although no such specific teaching is present in the reference. Appellant reiterates that what the reference may or may not "imply" is different from that which is "taught" by a reference. For a §103 rejection to be proper, the reference must teach or suggest every element of the claims. Suggested "implications" of a reference, as determined solely by the Examiner and without actual support in the reference, are not sufficient.

Furthermore, the Examiner's argument presupposes that one flame is the same as any other flame; according to the Examiner, any "flame" necessarily anticipates or renders obvious an "annular" flame. Appellants disagree. As described previously, Exhibit B1, Charojrochkul, et al.'s states at page 2517, second paragraph, that "Three different flame configurations were produced by changing the deposition parameters….The shape of the combustion zone was later found to have a **marked effect** on the microstructure of the deposited film." (Emphasis added).

That is, post-filing, peer-reviewed references have demonstrated that significant differences in the deposited material occurs when the flame configurations are altered. The shape of the flame, i.e., an annular flame vs. a continuous flame, is clearly applicable to the flame configuration, such that an alteration between the flame shapes would significantly alter the material deposition. One of skill in the art would certainly be aware of the relationship between flame configurations (including shape) and material deposition properties, and therefore would not attempt to alter the exact flame configuration of a reference, including the flame shape. Thus, one of skill in the art would not read a reference that **does not specifically describe** an annular flame as teaching or suggesting an annular flame.

The Examiner's Answer includes claims 52-56 in the §103 rejection over Choy et al. in view of Hitachi for the first time. *See* "Rejection 4" heading at page 12 of Examiner's Answer. However, the discussion of the rejection refers only to claims 29-44, and does not at all mention claims 52-56. Thus, Appellants assume that the inclusion of claims 52-56 in the rejection heading was in error. If the rejection is now to be applied to claims 52-56, Appellants request issuance of a revised Examiner's Answer to make such rejection clear.

As the pending claims require the presence of an annular flame and the Examiner has failed to specifically indicate where Hitachi teaches an annular flame, the applicant maintains that Hitachi does not disclose the generation of an annular flame combustion region as alleged by the Examiner, and therefore does not correct the defects of Choy *et al.*

E. Claim 51 is rejected under 35 U.S.C. §103(a) as unpatentable over Choy *et al.* (WO 97/21848) in view of Blackwell *et al.* <u>(U.S. Patent 6,312,656) and further in view of Masahide</u>

There is no prima facie obviousness between the instant claim and Choy et al .and Blackwell et al. in combination with JP 62-220376.

It is respectfully submitted that Choy *et al*. and Blackwell *et al.* in combination with Masahide does not render obvious the present invention of an apparatus for depositing material on a substrate, comprising: a nozzle assembly including a first outlet from which a stream of droplets of a precursor liquid is in use delivered to a substrate, and a second outlet from which a flow of fuel is in use delivered such as to provide an annular flame combustion region through which at least a portion of the stream of droplets in use passes before reaching the substrate; a precursor supply for supplying a precursor liquid to the nozzle assembly; an electrical supply for applying an electric field between the first outlet and the substrate; and a burner for generating the flame of the annular flame combustion region between the first outlet and the substrate; whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, in the annular flame combustion region to provide the deposited material.

All of the June 11, 2004, April 13, 2005 and December 27, 2005 Office Actions stated that it would have been obvious to use a wire-mesh electrode like that of Masahide in the arrangement of Choy *et al*. and Blackwell *et al*., because a mesh would have been equivalent to the ring electrode of Choy *et al*. Applicants have disagreed with this assertion in previous responses (the contents of which are provided herein) and continue to do so.

As argued in the October 13, 2005 Response, and as set forth herein, the mesh electrode of Masahide serves to create a corona discharge between two electrodes. This is in contrast to the present invention, which already contains an electric field which is generated from high voltage source 45, and exists between the nozzle assembly 10 and substrate 50. Rather than acting as the source of the electric field in the present invention, and indeed, the mesh of the present invention is not related to the presence of the electric field in any sense, the mesh instead serves to assist in removing soot from the flame.

Therefore, as the mesh in the present invention has a different function than that of the mesh in JP 62-220376, the Examiner's assertion that it would have been obvious to combine the mesh of Masahide with the arrangement Choy *et al.* and Blackwell *et al.* has no basis as there is no motivation for one of skill in the art to add a mesh electrode to an apparatus which already contains means for generating an electric field.

Again, the Examiner has repeatedly indicated that the cited reference "implies" the use of an annular flame, although no such specific teaching is present in the reference. Appellant reiterates that what the reference may or may not "imply" is different from that which is "taught" by a reference. For a §103 rejection to be proper, the reference must teach or suggest every element of the claims. Suggested "implications" of a reference, as determined solely by the Examiner and without actual support in the reference, are not sufficient.

Furthermore, the Examiner's argument presupposes that one flame is the same as any other flame; according to the Examiner, any "flame" necessarily anticipates or renders obvious an "annular" flame. Appellants disagree. As described previously, Exhibit B1, Charojrochkul, et al.'s states at page 2517, second paragraph, that "Three different flame configurations were produced by changing the deposition parameters….The shape of the combustion zone was later found to have a **marked effect** on the microstructure of the deposited film." (Emphasis added).

That is, post-filing, peer-reviewed references have demonstrated that significant differences in the deposited material occurs when the flame configurations are altered. The shape of the flame, i.e., an annular flame vs. a continuous flame, is clearly applicable to the flame configuration, such that an alteration between the flame shapes would significantly alter the material deposition. One of skill in the art would certainly be aware of the relationship between flame configurations (including shape) and material deposition properties, and therefore would not attempt to alter the exact flame configuration of a reference, including the flame

shape. Thus, one of skill in the art would not read a reference that **<u>does not specifically describe</u>** an annular flame as teaching or suggesting an annular flame.

Furthermore, as discussed in previous responses and above, Applicants maintain that the combination of Choy *et al*. and Blackwell *et al*. fail to teach an apparatus which generates an annular flame, and the combination of these documents with Masahide does nothing to remedy this deficiency.

In summary, it is submitted that a person skilled in the art would not have contemplated modifying the already deficient apparatus or method formed by combination of Choy *et al*. and Blackwell *et al*. in accordance with the teaching of Masahide to use a wire mesh electrode, and furthermore such a combination fails to teach an annular flame, and, notwithstanding any such modification of Choy *et al*. and Blackwell *et al*., the claimed invention is clearly distinguished over the disclosures of Choy *et al*., Blackwell *et al*. and Masahide when taken in combination.

CONCLUSION

For the reasons discussed in this brief and the arguments of record (which are both recited herein and incorporated herein by reference), claims 45-50 and 52-56 are patentable over Hitachi (JP 56-5337), claim 51 is patentable over Hitachi in view of Masahide, claims 29-56 are patentable over Choy *et al.* (WO 97/21848) in view of Blackwell *et al.* (US 6,312,656), claims 29-44 and 52-56 are patentable over Choy *et al.* (WO 97/21848) in view of Hitachi (JP 56-5337), and claim 51 is patentable over Choy *et al.* (WO 97/21848) in view of Blackwell *et al.* (U.S. Patent 6,312,656) and further in view of Masahide. It is, therefore, respectfully submitted that the Examiner erred in rejecting claims 29-56, and a reversal of the rejection of claims 29-56 by this Honorable Board, and prompt issuance of a Notice of Allowance, are earnestly solicited.

Respectfully submitted,

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CLAIMS APPENDIX A

Currently Pending Claims:

29. A method of depositing material on a substrate, comprising the steps of:

delivering from a first outlet a stream of droplets of a precursor liquid towards a substrate;

applying an electric field between the first outlet and the substrate; and

delivering from a second outlet a flow of fuel about the stream of droplets such as to provide an annular flame combustion region between the first outlet and the substrate through which at least a portion of the stream of droplets passes before reaching the substrate, whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, to provide the deposited material.

30. The method according to claim 29, wherein the chemical reaction, or the decomposition, or the chemical reaction and decomposition, occurs in a higher temperature overlap zone between the stream of droplets and the annular flame combustion region.

31. The method according to claim 29, wherein the flow of fuel is a diverging flow.

32. The method according to claim 29, wherein the first and second outlets are coaxial.

33. The method according to claim 29, wherein the stream of droplets is provided as a diverging spray.

34. The method according to claim 29, further comprising the step of:

delivering a flow of cold gas in a direction from the first outlet towards the substrate.

35. The method according to claim 34, wherein the flow of cold gas is delivered from a third outlet as a flow about the stream of droplets and within the flow of fuel.

36. The method according to claim 35, wherein the first and third outlets are coaxial.

37. The method according to claim 29, wherein the material is a ceramic material.

38. The method according to claim 29, wherein the material is a multicomponent oxide material.

39. The method according to claim 29, further comprising the step of:

heating the substrate.

40. The method according to claim 29, wherein the precursor liquid is a sol precursor solution.

41. The method according to claim 29, further comprising the step of:

moving one or both of the substrate and the first outlet during deposition so as to deposit a three-dimensional structure as a series of overlying layers.

42. The method according to claim 29, further comprising the step of:

controlling a region of deposition by varying one or more of a rate of flow of the fuel, a separation between the first outlet and the substrate and the electric field between the first outlet and the substrate.

43. The method according to claim 29, wherein the material is deposited as a powder and the chemical reaction, or the decomposition, or the chemical reaction and decompostion, occurs away from the substrate.

44. The method according to claim 29, wherein the material is deposited as a solid film and the chemical reaction, or the decomposition, or the chemical reaction and decomposition, occurs in the vicinity of the substrate.

45. An apparatus for depositing material on a substrate, comprising:

a nozzle assembly including a first outlet from which a stream of droplets of a precursor liquid is in use delivered to a substrate, and a second outlet from which a flow of fuel is in use delivered such as to provide an annular flame combustion region through which at least a portion of the stream of droplets in use passes before reaching the substrate;

a precursor supply for supplying a precursor liquid to the nozzle assembly;

an electrical supply for applying an electric field between the first outlet and the substrate; and

a burner for generating the flame of the annular flame combustion region between the first outlet and the substrate;

whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, in the annular flame combustion region to provide the deposited material.

46. The apparatus according to claim 45, wherein the chemical reaction, or the decomposition, or the chemical reaction and decompostition, occurs in a higher temperature overlap zone between the stream of droplets and the annular flame combustion region.

47. The apparatus according to claim 45, wherein the first and second outlets are coaxial.

48. The apparatus according to claim 45, wherein the nozzle assembly further comprises a third outlet disposed between the first and second outlets from which a flow of cold gas is in use delivered.

49. The apparatus according to claim 48, wherein the first and third outlets are coaxial.

50. The apparatus according to claim 45, wherein the first outlet is a central outlet.

51. An apparatus for depositing material on a substrate, comprising:

a nozzle assembly including a first outlet from which a stream of droplets of a precursor liquid is in use delivered to a substrate, and a second outlet from which a flow of fuel is in use delivered such as to provide an annular flame combustion region through which at least a portion of the stream of droplets in use passes before reaching the substrate;

a mesh disposed between the first outlet and the substrate;

a precursor supply for supplying a precursor liquid to the nozzle assembly;

an electrical supply for applying an electric field between the first outlet and the substrate; and

a burner for generating the flame of the annular flame combustion region between the first outlet and the substrate;

whereby the precursor liquid is chemically reacted, or decomposed, or chemically reacted and decomposed, in the annular flame combustion region to provide the deposited material.

52. The apparatus according to claim 45, further comprising:

an electrode at an electric potential between the potential of the first outlet and the substrate and disposed between the first outlet and the substrate.

53. The apparatus according to claim 52, wherein the electrode is an annular electrode.

54. The apparatus according to claim 45, further comprising:

a positioner for altering the relative position of the first outlet and the substrate.

55. The apparatus according to claim 45, where configured such that the chemical reaction, or the decomposition, or the chemical reaction and decomposition, occurs away from the substrate so as to provide the material as a powder.

56. The apparatus according to claim 45, where configured such that the chemical reaction, or the decomposition, or the chemical reaction and decomposition, occurs in the vicinity of the substrate so as to provide the material as a solid film.

EVIDENCE APPENDIX B

B1. Charojrochkul, et al., "Flame assisted vapour deposition of cathode for solid oxide fuel cells. 1. Microstructure control from processing parameters." Journal of the European Ceramic Society, 2004, 24:2515-2526.

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Flame assisted vapour deposition of cathode for solid oxide fuel cells. 1. Microstructure control from processing parameters

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Abstract

The flame assisted vapour deposition (FAVD) technique has been used to deposit porous lanthanum strontium manganese oxide (LSM) to produce the cathode for a solid oxide fuel cell (SOFC). FAVD combines flame synthesis and vapour deposition methods where the liquid precursor undergoes a combustion process into a vapour phase, and then deposits as an oxide film on a substrate. The work has shown that the microstructures of deposited films may be dense or porous depending on processing parameters such as the deposition temperature, the concentration of the fuel and the distance between the spray nozzle and the substrate. A mechanism for the effects of these processing parameters interpreted as 'the combustion zone' has been proposed. This mechanism is used to explain the physical properties of films which were characterised using SEM and XRD. The FAVD technique is shown to be an efficient way in which SOFCs' cathode film can be fabricated with tailoring of the desired phases and microstructure.

Keywords: Cathodes; Flame assisted vapour deposition; Fuel oils; (La,Sr)MnO_3; SOFC

1. Introduction

Solid oxide fuel cells (SOFC) are being developed as efficient power generators. Difficulties involving the fabrication of the cell components, materials selection and high operating temperatures associated with SOFCs still have to be overcome before this technology becomes commercially viable. The present work involves the development of SOFC components fabrication, especially the cathode.

SOFC components can be fabricated via routes such as electrochemical vapour deposition (EVD) which is used commercially,[1] Chemical Vapour Deposition (CVD),[2] and Physical Vapour Deposition (PVD).[3] A novel method of Flame Assisted Vapour Deposition (FAVD) has been proposed to deposit a porous cathode film for SOFCs. This method has a similar principle to conventional CVD in which a film is deposited from the vapour phase.[4] Preliminary work showed the possibility of using this technique to deposit porous films of $La_{1-X}Sr_XMnO_3$ (LSM).[5–7] It is a cost effective method which yields a film of reasonable quality both in terms of microstructure and electrical properties.[6,7] The microstructure of LSM is required to be porous for the oxygen diffusion within the cathode to the three phase boundary between gas/cathode/electrolyte to take place.[8] The aim of the current research is to study the effect of processing parameters on the microstructure and phases present in deposited films. Consequently, some processing parameters such as the amount of fuel and the effect of droplet size (from the air pressure variation) have been modelled using the Computational Fluid Dynamic software package FLUENT. The results obtained from this model have been used to support the experimental results. The detail of the modelling work is described in a subsequent paper.[9]

2. Experimental procedures and setup

2.1. Preparation of a dense YSZ electrolyte substrate

An 8 mol% Y_2O_3 stabilised ZrO_2–YSZ was selected as the SOFC electrolyte. Commercial YSZ powder

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(supplied by Tosoh, Japan) was pressed into green body pellets of 10 mm in diameter by around 0.5–0.7 mm in thickness. The pellets were then sintered in air at 1500 °C for 2 h to produce densities greater than 98%.

2.2. Preparation of the precursor solution

Metal nitrate compounds of La [99.9% $La(NO_3)_3xH_2O$], Sr [99+% $Sr(NO_3)$, and Mn (98% $Mn(NO_3)_26H_2O$] supplied by Aldrich Chemicals, were dissolved in deionised water to the desired ratio of La:Sr:Mn = 0.82:0.18:1 and 0.61:0.18:1. Ethanol was added to increase the inflammability of the solution. The ratio of ethanol to water was varied from 50:50 to 80:20 while the concentration of the precursor was varied from 0.01 to 0.1M.

2.3. Flame Assisted Vapour Deposition (FAVD) set up

A dense YSZ substrate was heated by means of a hot plate and a flame torch (such as a bunsen burner). The precursor solution as prepared previously was sprayed through an air atomiser across the flame to the substrate situated vertically below the spray nozzle. The atomised precursor was in the form of fine droplets, which are assumed to be in the form of an aerosol. These droplets then combusted when ignited by a flame source. The deposition temperature was monitored using a thermocouple placed next to the substrate. The heat of combustion provided thermal energy during the decomposition and vapourisation of these droplets, which underwent vapour phase reactions, resulting in an oxide film being deposited on a substrate. The full details of FAVD are described in previous literature.[4–6] The effects of processing parameters were investigated by varying only one parameter at a time while the others were kept constant. The full range of experimental conditions are given in Table 1.

3. Characterisation techniques

The adhesion of the as deposited films was evaluated qualitatively using a simple scratch test with an adhesive tape. The deposition was considered powdery when some particles remained on the adhesive tape. The microstructures of the films were examined using scanning electron microscope (SEM). Phases present in the films were identified by X-ray diffraction using Theta-2 Theta setup (Philips PW1710, with Cu target).

4. Results and discussion

4.1. Effect of ethanol/water ratio in the precursor solution

The ratios of ethanol/water in the precursor solution were varied as stated in Table 1. These variations produced changes in the deposition temperature and the characteristics of the as-deposited films. The high ethanol/water ratio resulted in a high deposition temperature while the low ethanol/water ratio gave low deposition temperature. At 770–810 °C, the deposition temperature of the 80/20 ratio produced film with powder agglomeration as shown in Fig. 1(a). However, the powder produced was very fine, ~1 micron in diameter and could be removed easily. A uniform porous film of large particles (10–20 microns size) was obtained at the 70/30 ratio (650–690 °C) as in Fig. 1(b). In Fig. 1(c), the film was nearly dense but with some large particles from the 60/40 ratio deposited at 470–520 °C. SEM micrographs on the right reveal the cross-section of the deposited film where the film thickness can be estimated.

The XRD spectra of the 70/30 ratio sample showed sharp crystalline peaks of $La_{0.8}Sr_{0.2}MnO_3$ (JCPDF 40-1100) designated as LSM while the spectra of the 60/40 ratio showed only a few peaks of LSM on broad haloes

Table 1
Standard parameters and variations used in FAVD for each examining effect

Parameters variation	Conc. of precursor (Molar)	Ratio of ethanol/water	Flow rate of precursor (ml/min)	Distance between spray nozzle and substrate (cm)	Pressure of compressed air (psi)
Effect of ethanol/water ratio[a]	0.05	80/20, 70/30, 60/40	12	12	22
Effect of flow rate[b]	0.05	80/20	5.5, 10, 17	12	22
Effect of conc. of precursor[a]	0.1, 0.075, 0.05, 0.025, 0.01	70/30	12	12	22
Effect of pressure of air atomizer[a]	0.05	70/30	12	12	17, 22, 26
Effect of distance between nozzle and substrate[b]	0.05	80/20	12	8, 12, 16	22

[a] The ratio of La:Sr:Mn was 0.82:0.18:1.

(indicating amorphous structures) as shown in Fig. 2. No peaks were observed for any films deposited at lower ratio than 60/40 and lower deposition temperature. The result implied non-crystalline deposition.

The shapes of the flame during deposition (combustion of precursor solution) was observed and are shown schematically in Fig. 3. Three different flame configurations were produced by changing the deposition parameters: these are designated as combustion zone A, B, and C. The area of zone A is the least while zone B includes zone A, and zone C includes both zones A and B. The shape of the combustion zone was later found to have a marked effect on the microstructure of the deposited film. The effect of the thermal environments was reported by Wiedmann et al.[4] and Viguie et al.[10]

At a high ethanol ratio (80/20), the range of deposition temperature was high since ethanol acted as a fuel for combustion resulting in a combustion zone of *Shape C* as seen in Fig. 3. By the time all the combusting species reached the substrate, the temperature was high, since the combustion process still continued. The difference of temperatures in the combustion and the sub-

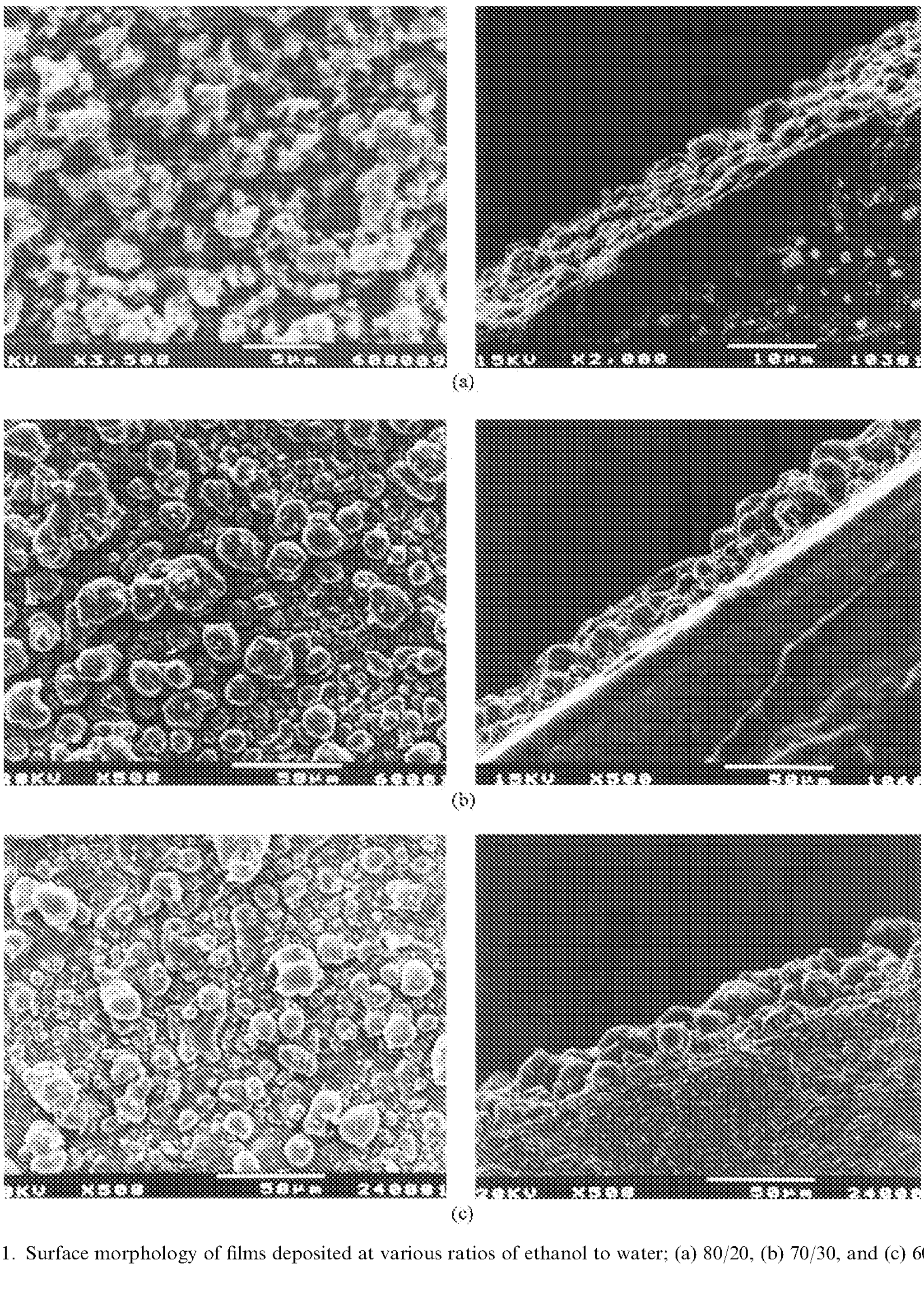

Fig. 1. Surface morphology of films deposited at various ratios of ethanol to water; (a) 80/20, (b) 70/30, and (c) 60/40.

strate was fairly low. Since the ratio of ethanol/water was high, the fuel was rich, most species combusted and reacted readily before reaching the substrate. As a result, the deposited particles were very fine as in Fig. 1(a) producing a powdery film. At high substrate temperature (700 °C), all the chemical reaction took place in the gas phase, in which the process was termed homogeneous gas phase nucleation.

When the ratio of ethanol/water was low enough (70/30) to yield the combustion zone of *Shape* B, the

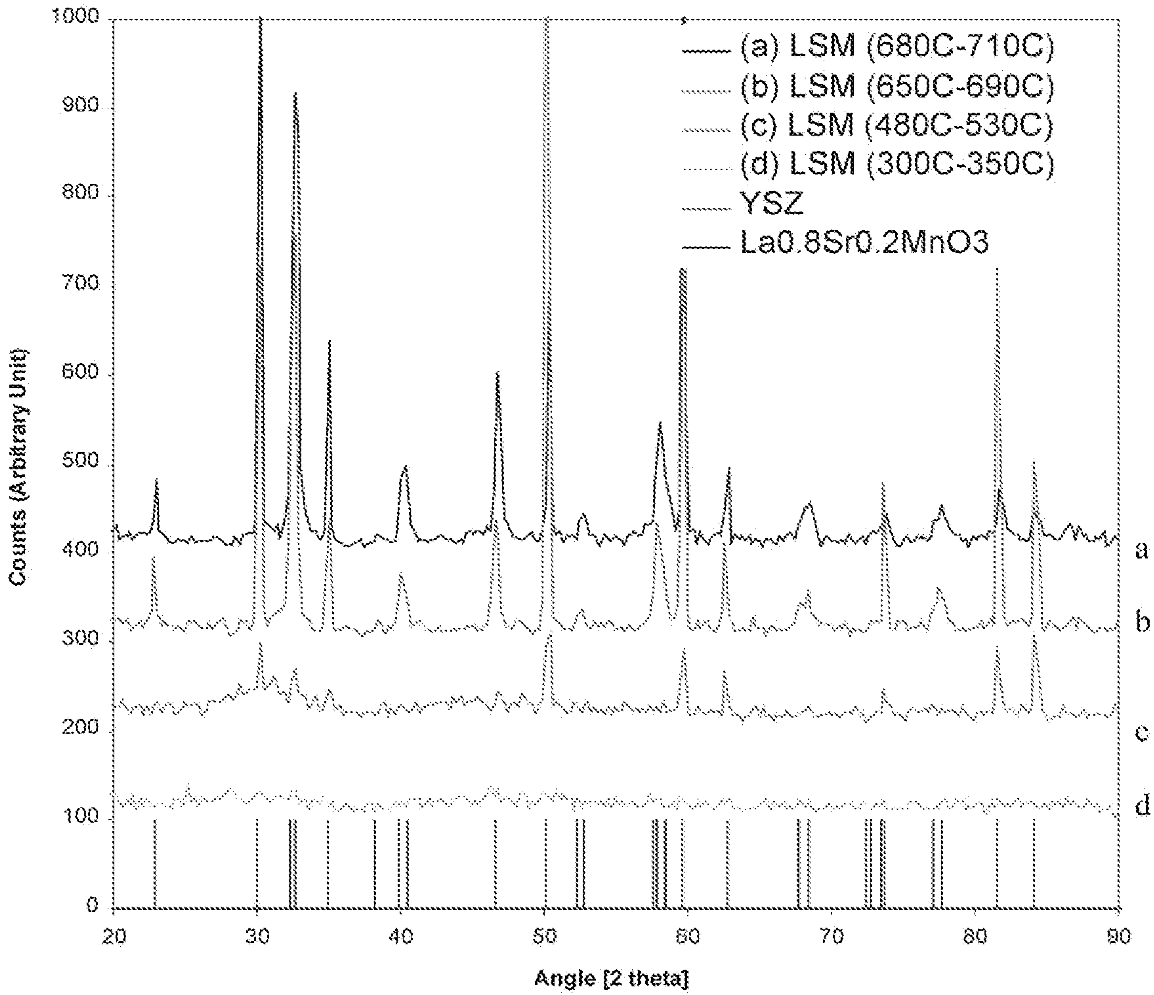

Fig. 2. XRD patterns of LSM films deposited at (a) 680–710 °C, (b) 650–690 °C, (c) 480–530 °C, and (d) 300–350 °C.

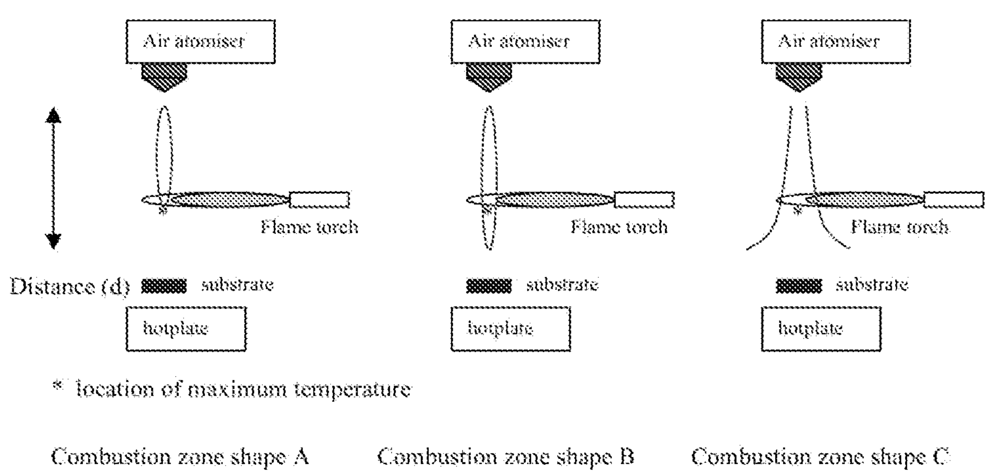

Fig. 3. A sketch of various combustion zones in the FAVD method.

deposition temperature was reduced. The heterogeneous nucleation reaction occured at the vicinity of the substrate resulting in a dense film as in Fig. 1(c). The maximum temperatures in the combustion zone of *Shape B* and *Shape C* were approximately the same, which was around 1000 °C. The maximum temperature at the tip of the flame torch was around 1200 °C. However, the heat convected/radiated to the substrate of combustion zone *Shape B* was lower than that of *Shape C* from the greater distance between the combustion and the substrate. In *Shape C*, the combustion was not complete within the combustion zone, therefore some aerosol droplets agglomerated and deposited as a porous film. During the deposition, particles grew and formed a densely packed film as in Fig. 1(b).

The combustion zone of *Shape A* only occured when the ratio of ethanol/water was very low (below 60/40) in that the fuel was not sufficient to combust all the droplets. The droplets then splashed onto the heated substrate. The solvent evaporated while leaving traces of dry precipitate, producing a flaky, amorphous film (Fig. 2(d)) as the decomposition of fuel and ions was not complete. The deposition temperature was very close to the temperature at the hot plate. The mechanisms are summarised in Fig. 4. The black open circle, grey open circle and black dot indicate an aerosol droplet, the reaction zone and the deposited particle respectively.

4.2. *Effect of flow rate of the precursor solution*

The flow rate of the precursor solution was varied from 5.5 ml/min to 17 ml/min whilst the deposition temperatures were controlled within the range of 700–800 °C. The microstructure of films deposited at the flow rate 5.5 ml/min, 10 ml/min, and 17 ml/min respectively are shown in Fig. 5.

At the low flow rate (5.5 ml/min), the combustion and decomposition reactions were completed in the gas phase, therefore the deposited film was powdery (the combustion zone of *Shape C*). At the high flow rate (17 ml/min), the reactions have not been completed by the time the droplets reached the substrate but continued to react at the vicinity of the substrate, resulting in a relatively dense film (the combustion zone of *Shape B*). Nevertheless, some cracks were present in the dense film probably due to thermal stresses on the thin dense film (around 4 microns thick). This dense film was not as resistant to cracking as a porous film. At the medium flow rate (10 ml/min), the film was a combination of particles and dense film resulting in a porous film.

The XRD patterns in Fig. 6 of various flow rate depositions showed the difference mainly for the substrate peaks. The film produced from the high flow rate (17 ml/min) deposition was much denser than the powdery film produced at a lower flow rate (5.5 ml/min).

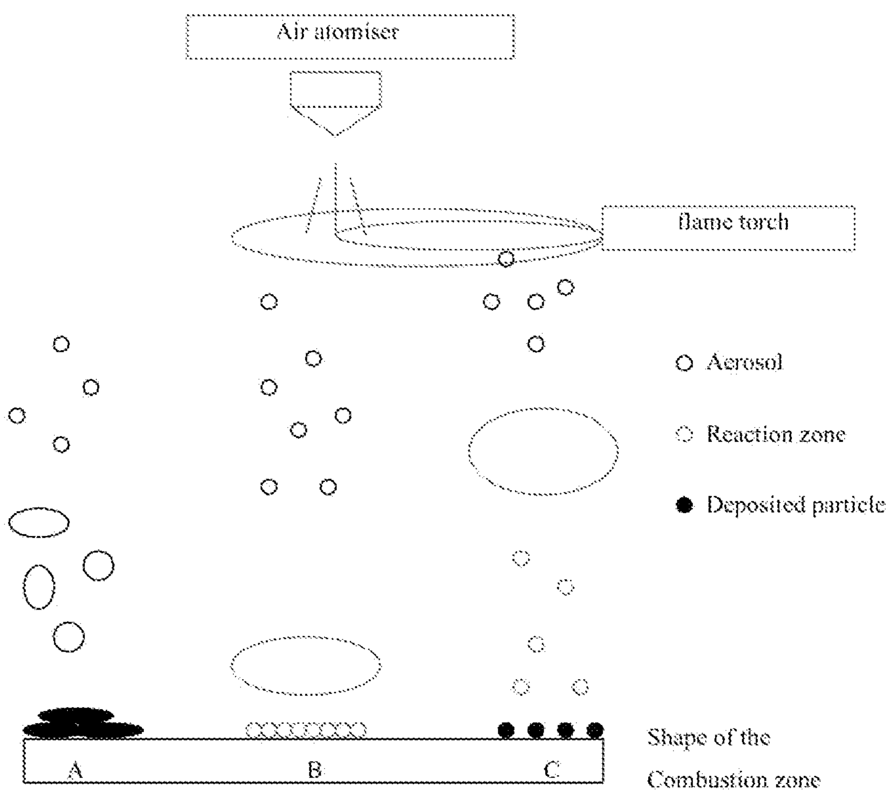

Fig. 4. A sketch of the relationship between the combustion zone and the deposition process.

The substrate peaks were more easily detected (referring to a thinner layer) and the powdery layer was more crystalline than the film deposited at the high flow rate. The LSM peaks were very similar in the shapes and degree of crystallinity.

4.3. Effect of concentration of the precursor solution

Fig. 6 shows the microstructures of the deposited films at various concentrations. The sizes of particles in the films varied with the concentration when they were deposited at the same ratio of ethanol/water. From the SEM micrographs, the size of particles was largest at high concentration (0.1 M) and smallest at low concentration (0.01 M). The degree of agglomeration also depended on the concentration of the precursor. More agglomeration was obtained at high precursor concentration, which also produced large particles as in Fig. 6(a). The porosity of the film was influenced by these parameters to form a large particle size with a high degree of agglomeration, associated with a high porosity fraction. The films showed less degree of agglomeration at reduced concentration when individual and finer particles became more distinctive. The film was the most powdery at the least concentration as seen in Fig. 6(e).

The effect of precursor concentration on microstructure can be described as the compaction of particles. When individual particles were close to each other from high concentration solution, they agglomerated. At low concentration, particles were far from each other and deposited as individual particles.

A similar influence has also been observed by Zhang and Messing[11] in using a spray pyrolysis technique to produce zirconia particles, the size of particle formed depended on the concentration of the solution. The size of particles was smaller when the concentration of solution was reduced.

4.4. Effect of the air atomiser pressure

The range of pressure variation of between 17 and 30 psi was restricted by the experimental setup. Fig. 7 shows large variations in the microstructure with the changes in compressed air pressures in the atomiser. The deposition temperature was controlled by the combination of all the parameters. At low pressure (17 psi), the film was powdery with fine particle sizes, smaller than 2 microns when the deposition temperature was at 720–770 °C. Particles in the film became connected at higher pressure as in Fig. 7(b) at 22 psi and the deposi-

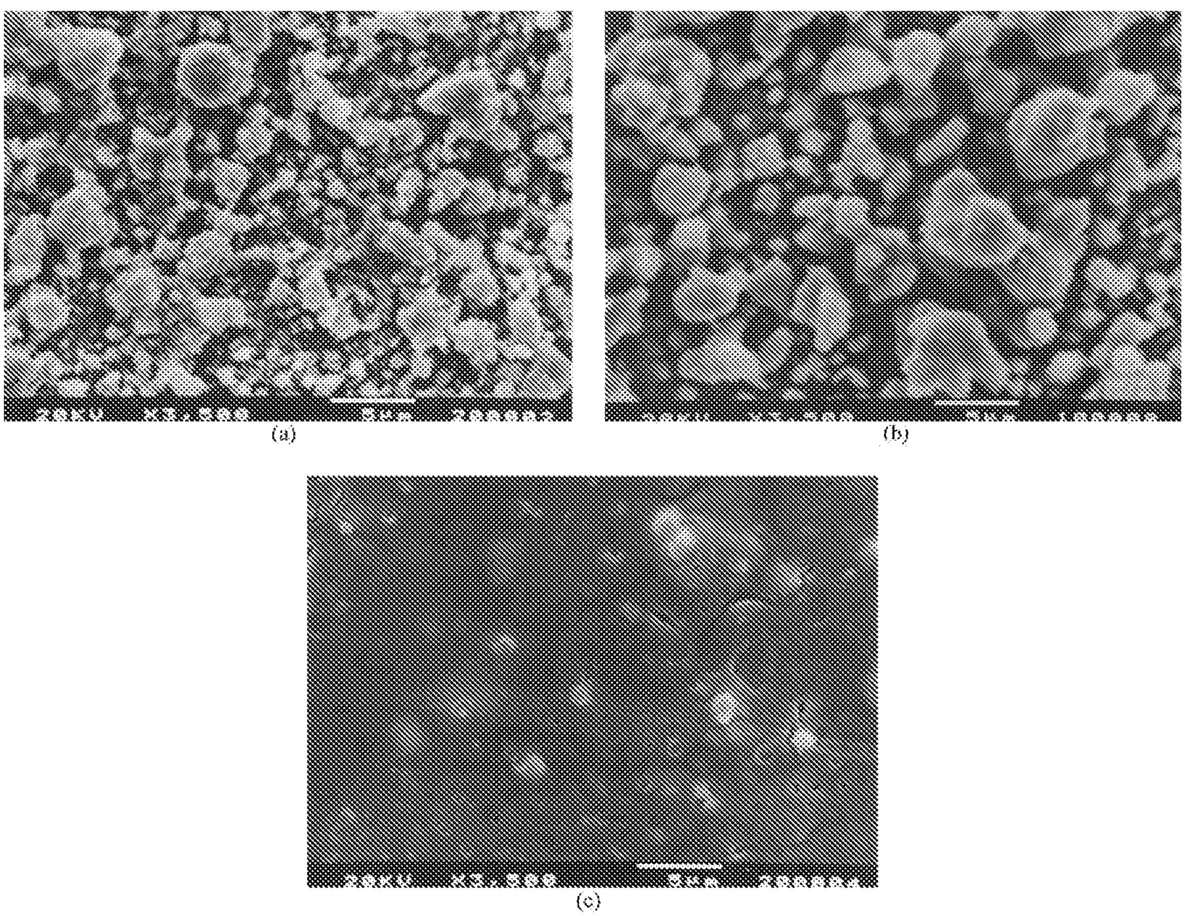

Fig. 5. Surface morphology of films deposited at (a) 5.5 ml/min, (b) 10 ml/min, and (c) 17 ml/min.

tion temperature was reduced to 680–730 °C. When the air pressure was increased further, the film was more densely packed with larger particle sizes of up to 20 microns in Fig. 7(c). However, some cracks were observed due to high air pressure producing high flow rate of cold air to chill the deposited surface. The deposition temperature was lowered to 450–500 °C.

The XRD patterns in Fig. 8 show that the crystallinity of deposited films was seen to be similar. The substrate peaks from the deposition at 17 psi were more obvious, due to the film being a thin powdery layer. The LSM film deposited at 26 psi was not electrically conducting and the associated XRD indicated an amorphous phase. After annealing at 900 °C for 2 h, this film became crystalline and electrically conducting. The film deposited at 17 psi showed traces of $La_2Zr_2O_7$ phase. This reaction product is undesirable for the SOFC cathode. However, it is commonly detected for low Sr dopant

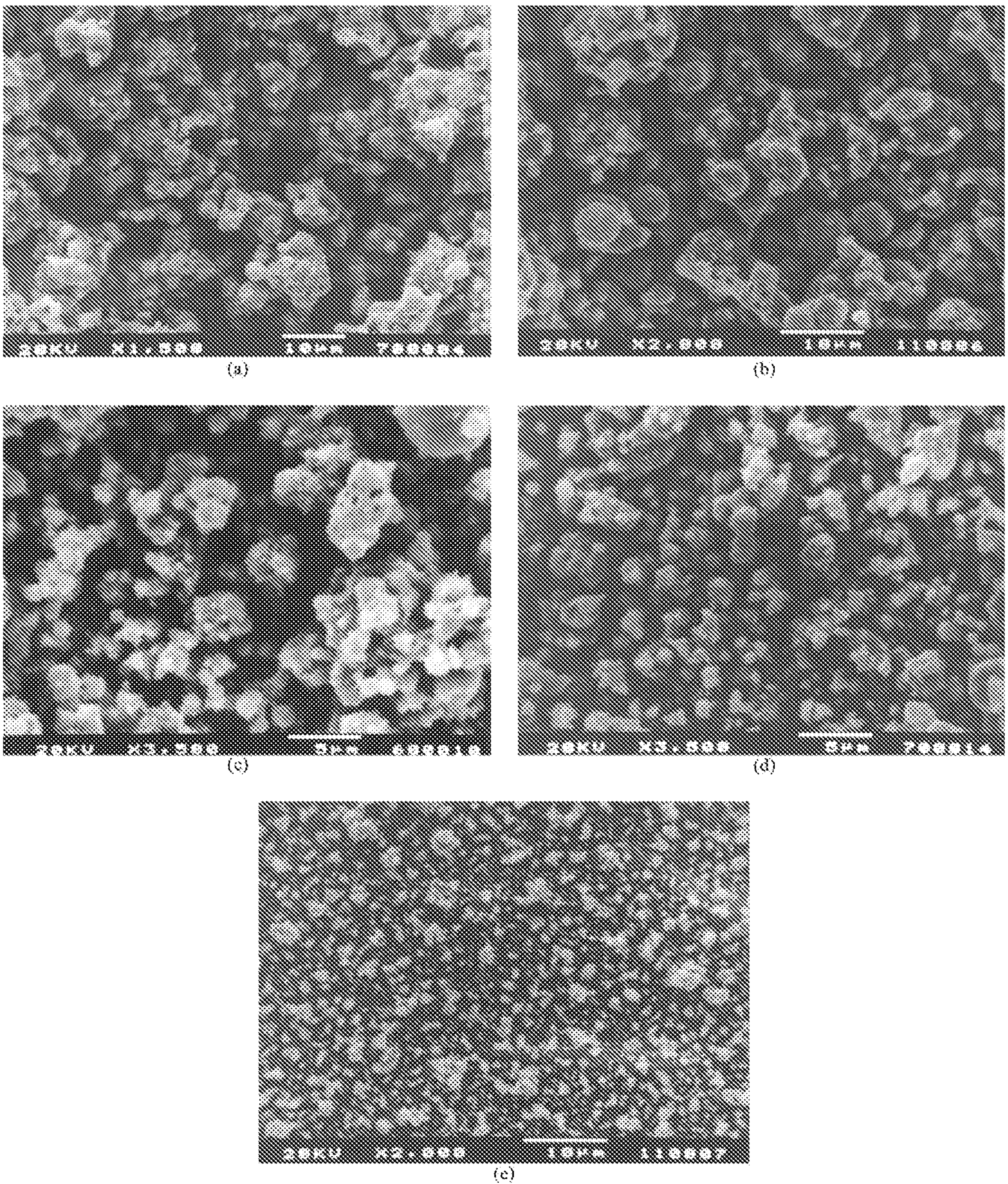

Fig. 6. Surface morphology of $La_{0.82}Sr_{0.18}MnO_3$ deposited from ethanol/water ratio of 70/30 of (a) 0.1 M, (b) 0.075 M, (c) 0.05 M, (d) 0.025 M, and (e) 0.01 M.

concentration as manganese diffuses to YSZ and the remaining lanthanum reacts with YSZ. $La_2Zr_2O_7$ has poor electrical conductivity and the coefficient of thermal expansion is not compatible with that of YSZ.[12–14] This acts as an insulating layer and creates thermal stresses at the interface.[15]

The total mass flow was the sum of the precursor solution and the compressed air. When the compressed air pressure was reduced, the amount of compressed air in the flow decreased, but it raised the ratio of the precursor solution in the total mass flow. As a result, the deposition temperature was increased in the higher fuel for combustion (higher ratio of solution to air). The compressed air was at room temperature therefore, it was acting as a stream of cool air and hence lowered the deposition temperature dramatically when the compressed air pressure was increased. The effect of deposition temperatures on the microstructures of the films was similar to the effect of ethanol/water ratio in the precursor solution. Fig. 9 shows a sketch of deposition processes at various compressed air pressures. At low air pressure, the deposition process followed the flame pattern of combustion zone of *Shape C* which yielded powdery film. At high air pressure, the flame pattern of this deposition process was similar to the combination of combustion zone of *Shapes B* and *C* producing a deposit of agglomerated particles.

4.5. Effect of the distance between the air atomiser and the substrate

The distance between the air atomiser and the substrate (*d*) was varied within the range of 8–16 cm as the shorter or longer distances had proven to be impractical. The surface morphology of the films produced at different distances are shown in Fig. 10. At a distance of 8 cm, the deposition temperature fluctuated tremendously from 500 to 830 °C resulting in a smooth film with fine pores and a lot of large cracks [Fig. 10 (a)]. The adhesion of the coating to the substrate was also very poor. At 12 cm, the film was uniformly porous with some agglomeration as in Fig. 10(b) and the normal range of deposition temperature (700–800 °C) was observed. At the longest distance of 16 cm, the deposition temperature varied widely again from 560 to 730 °C. The film produced was very powdery as in Fig. 10(c).

The $La_{0.8}Sr_{0.2}MnO_3$–LSM (JCPDF 40-110) phase was detected in all deposited films however the traces of

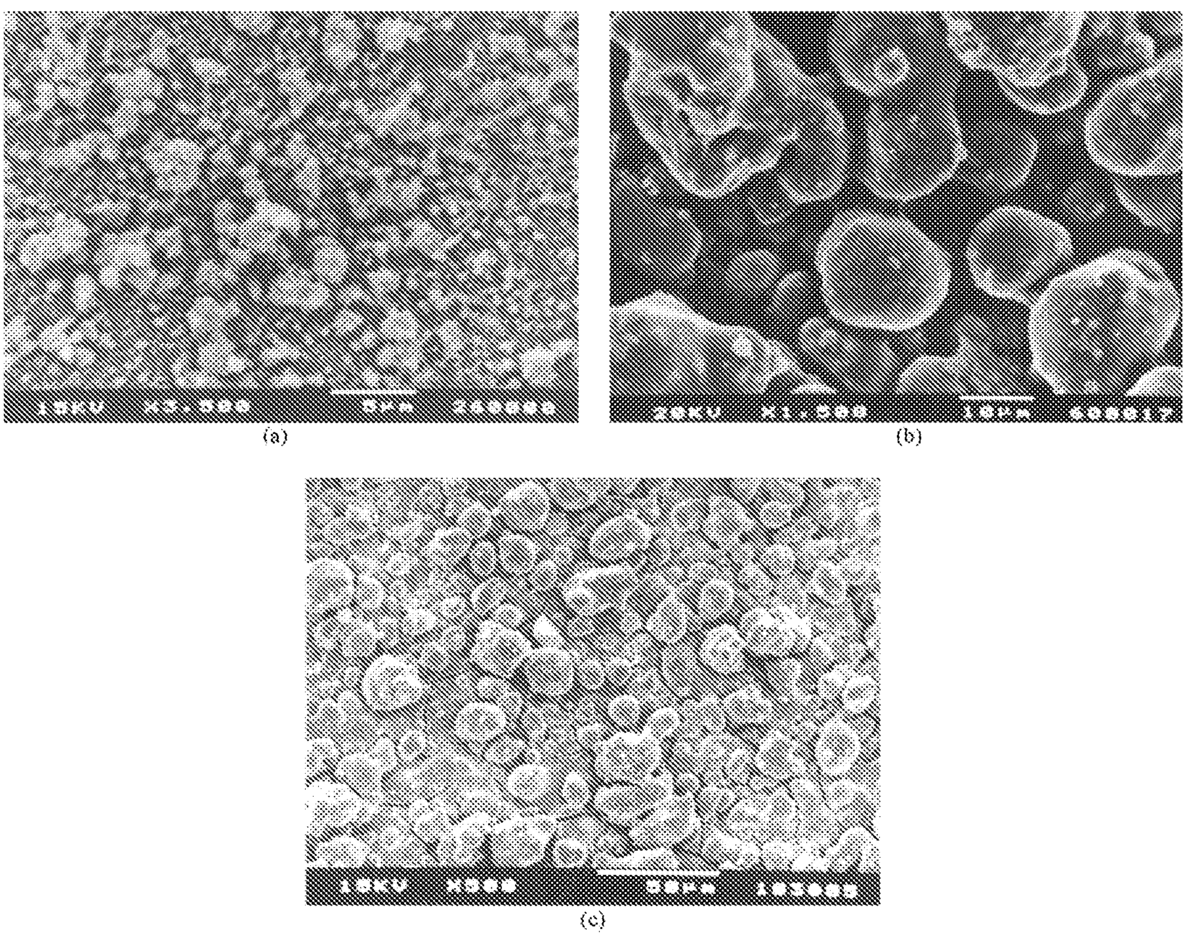

Fig. 7. Surface morphology of $La_{0.82}Sr_{0.18}MnO_3$ films deposited at (a) 17 psi, (b) 22 psi, and (c) 26 psi.

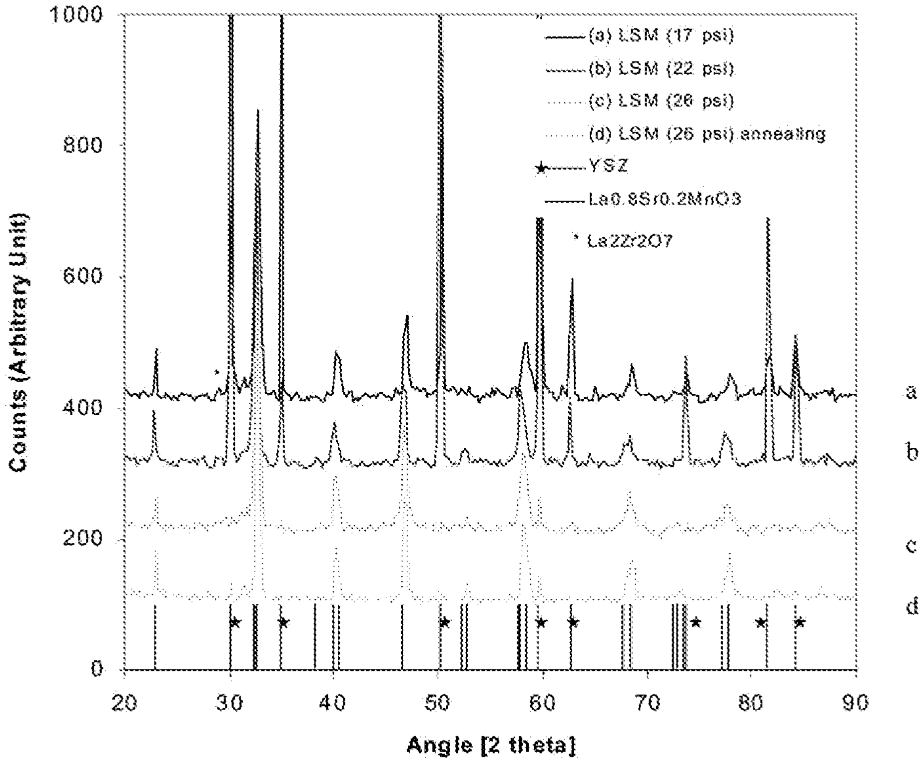

Fig. 8. XRD patterns of LSM films deposited at (a) 17 psi, (b) 22 psi, (c) 26 psi, and (d) 26 psi after annealing.

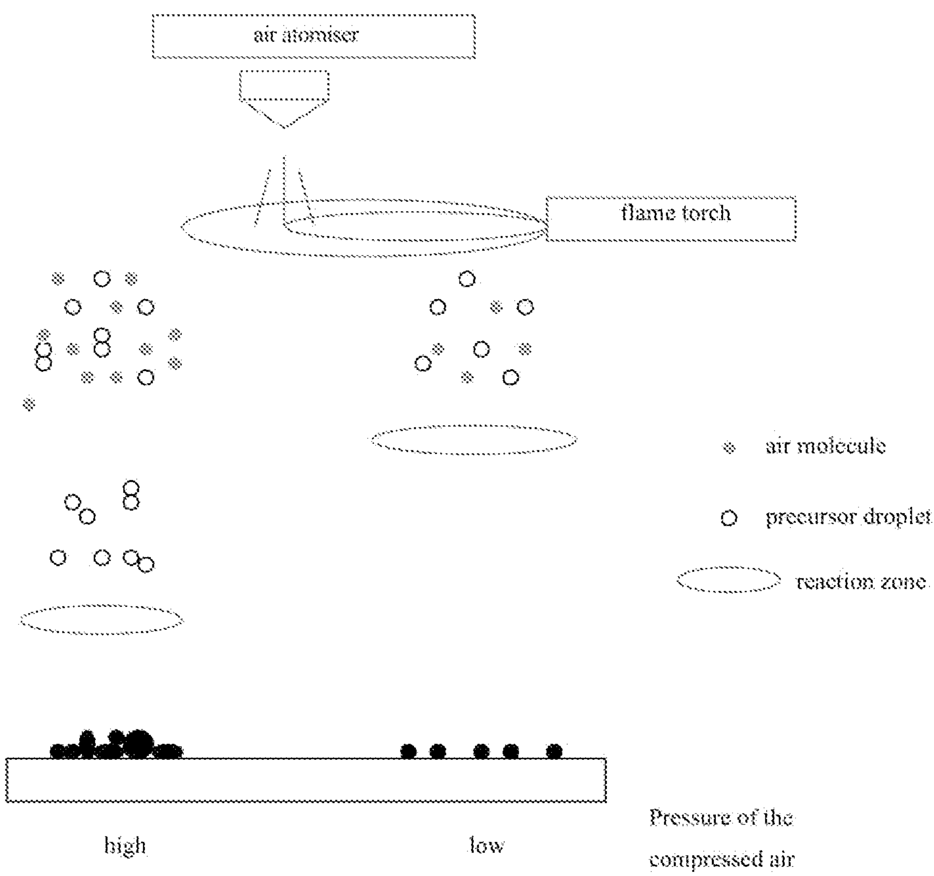

Fig. 9. A sketch of deposition processes at various compressed air pressures.

impurities found were different. The XRD pattern of the film from distance 8 cm indicated an amorphous phase and LSM phase of low degree of crystallinity, while the pattern of longer distances showed more crystalline films. Peaks representing $La_2Zr_2O_7$ phase were detected in the pattern for the distance 16 cm film. These two patterns are displayed in Fig. 11. In conclusion, both distances are not suitable for a SOFC

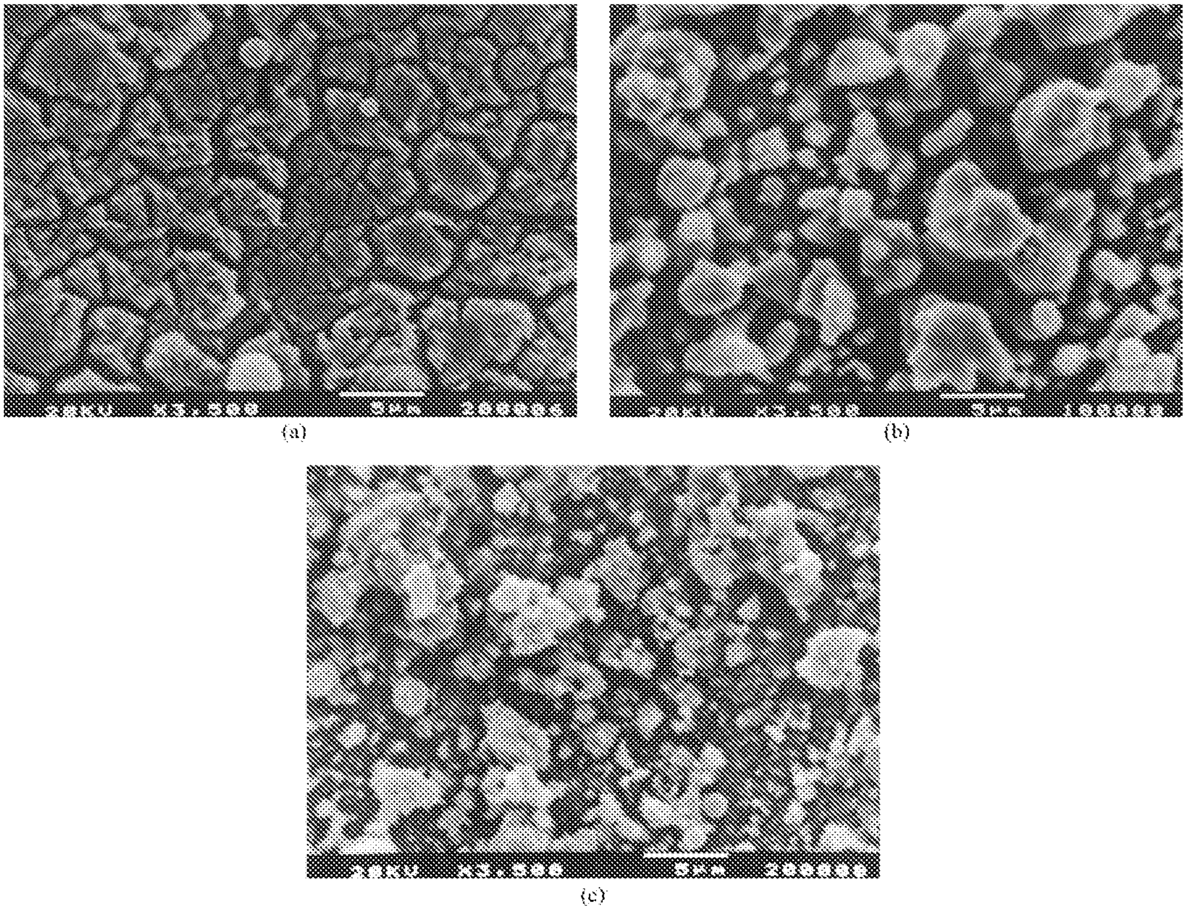

Fig. 10. Surface morphology of $La_{0.61}Sr_{0.18}MnO_3$ films deposited at distance (a) 8 cm, (b) 12 cm, and (c) 16 cm between the spray nozzle and the substrate.

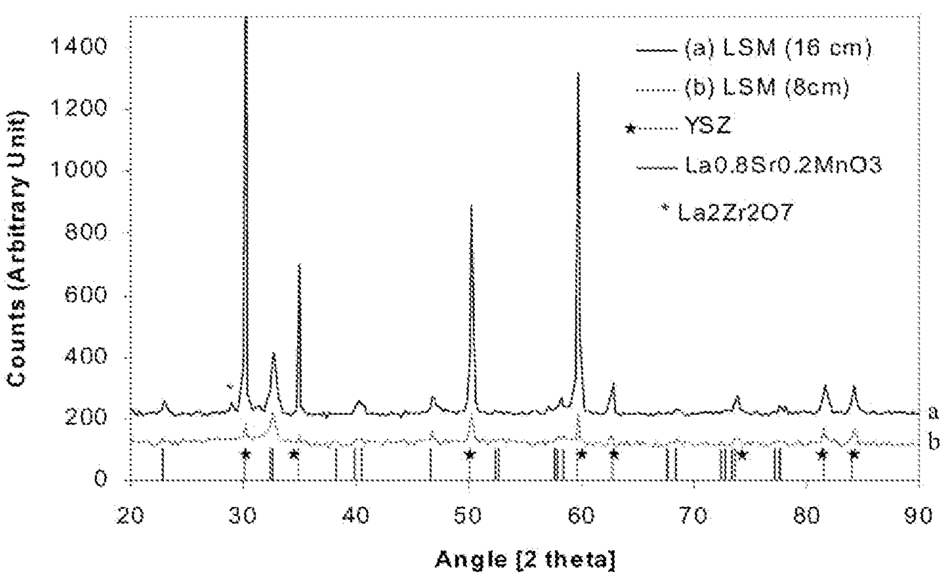

Fig. 11. XRD patterns of films deposited at distance (a) 16 cm and (b) 8 cm between the spray nozzle and the substrate.

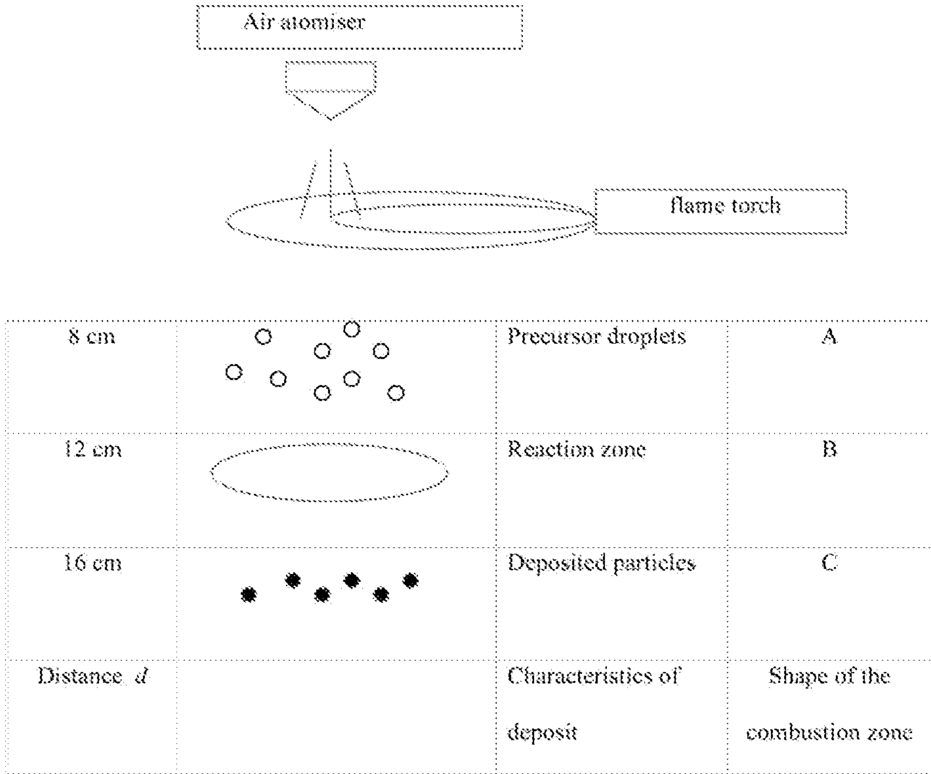

Fig. 12. A sketch of the deposition process at various distances in the combustion zone.

cathode application. The most applicable distance is at 12 cm which is in between with combination of crystallinity and without the second phase.

At the distance 8 cm, the substrate was very close to the combustion zone and as a result, the droplets were deposited as a dense film. This was because the combustion and decomposition reactions took place at or near the surface of the substrate. Nevertheless, when the distance was too low, cracks were obvious from thermal shock effects due to pulses from the precursor pump. When d was at 16 cm, the combustion process was complete prior to reaching the substrate. The dominant reaction was the homogenous gas phase nucleation in which all the particles were produced prior to reaching the substrate. Moreover, by the time the droplets reached the substrate, the momentum was reduced and the particles did not pack, resulting in a powdery surface. The relationship between d and the deposition process in the combustion zone is shown in Fig. 12.

5. Conclusions

In FAVD, films deposited at high temperature (700–800 °C) were more powdery and crystalline than those from lower temperature deposition. The dominant reaction at high temperature deposition was governed by the gas phase nucleation. The temperature can be raised either by increasing the amount of ethanol in the precursor solution or increasing the mass fraction of ethanol, or adjusting the position of the flame torch. At lower temperatures where heterogeneous reactions were dominant, the decomposition and combustion reactions occured in the gas phase at or near the heated substrate, which resulted in dense film.

A desired porous LSM cathode film could be deposited using the following condition: concentration of 0.05 M of the precursor, the ratio of ethanol/water at 75/25, 12 ml/min flow rate, 12 cm distance between the spray nozzle and the substrate and the pressure of the compressed air fixed at 22 psi. The required phases and amount of porosity are necessary for the reduction of oxygen at the cathode which leads to a better performance of SOFCs.

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RELATED PROCEEDING APPENDIX C

None.